

# Photon antibunching in the fluorescence of individual colored centers in diamond

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We have observed photon antibunching in the fluorescence light emitted from a single N-V center in diamond at room temperature. The possibility of generating triggerable single photons with such a solid state system is discussed.

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The security of quantum cryptography is based on the fact that each bit of information is coded on a single quantum object, namely a single photon. The fundamental impossibility of duplicating the complete quantum state of a single particle prevents any potential eavesdropper from intercepting the message without the receiver noticing<sup>1</sup>. In this context, the realization of a efficient and integrable light source delivering a train of pulses containing one and only one photon appears as an extremely challenging goal<sup>2</sup>. For reaching it, many issues should be addressed, which range from achieving the full control of the quantum properties of the source to an easy handling and integrability in a practical quantum cryptography set-up.

Several pioneering experiments have been realized for obtaining single photon sources<sup>3-7</sup>. Refs<sup>3,4</sup> are based on the spontaneous emission of twin photons. One of the photon is used to open a time gate during which the twin photon can be observed. In these types of set-ups, the emission time of the photon is random. In quantum cryptography based communication systems it is desirable to deal with single photons synchronized on an external clock, namely triggerable single photons<sup>5-7</sup>. Experiment by Kim et al<sup>6</sup> uses the regulation of the electron flux at the level of a single electron in a light emitting mesoscopic p-n junction to realize a single photon turnstile device. Experiments by de Martini et al<sup>5</sup> and Brunel et al<sup>7</sup> are based on the collection of fluorescence light emitted by a single dye molecule. The experiments reported in<sup>6,7</sup> require cryogenic apparatus (liquid He temperature). The experiment reported in<sup>5</sup> may run at room temperature, but then the molecules are rapidly destroyed by photobleaching and must be renewed, e.g. by having them in a liquid solvent<sup>8</sup>. Another general limitation in these experiments is the very small collection efficiency ( $\eta < 10^{-3}$ ), which means that less than 0.1% of the emitted photons are actually detected.

Generally speaking, the physical principle of single-molecule sources is that a single emitting dipole will emit only one photon at a time. Using an adequate pulsed excitation scheme, it is then possible to produce only one photon per pulse<sup>5,9</sup>. In this letter, we report on the use of single colored centers in diamond as single quantum emitters<sup>10</sup>. By investigating the quantum statistics of the fluorescence light we have observed photon antibunching, which is a clear signature of the unicity of the emitting dipole. Photon antibunching has already been observed in many experiments, involving e.g. a single atom<sup>11</sup>, a single trapped ion<sup>12</sup>, and a single molecule<sup>13</sup>. With respect to these experiments, we point out that our set-up is particularly simple, since it involves bulk diamond at room temperature, and a non-resonant excitation from an argon-ion laser at 514 nm, with a typical power of 10 mW. The present collection efficiency is still low (see discussion below), but straightforward improvements are possible. We consider thus that the present result is a first step towards the realization of a simple and efficient solid state source for single photons.

The colored center that has been used is the Nitrogen-Vacancy (N-V) defect center in diamond, with a zero phonon line at a wavelength of 637nm. The defect consists in a substitutionnal nitrogen and a vacancy in a adjacent site. A simplified level structure is a four-level scheme with fast non radiative decays within the two upper states and within the two lower states. The excited state lifetime is  $\tau = 11.6$  ns<sup>14</sup>. We have used  $0.1 \times 1.5 \times 1.5$ mm<sup>3</sup> single [110] crystals of synthetic Ib diamond from Drukker International. Nitrogen is present in the crystal as an impurity. Vacancies are created by irradiation with 2 MeV electrons at a dose of  $3 \times 10^{12} e^- / \text{cm}^2$ . The irradiation dose is chosen so that the density of vacancies is of the order of  $1 \mu\text{m}^{-3}$ . After irradiation the crystals are annealed in vacuum at 850°C during 2 hours to form the N-V centers. A remarkable property of these centers is that we could not observe any photobleaching : the fluorescence level remains unchanged after several hours of continuous laser irradiation of a single center in the saturation regime. On the other hand, a limitation of the system is the existence of shelving in a metastable singlet state. This leads to the observation of photon bunching for time scale longer than the lifetime

of the center<sup>8,13,15</sup>, and to a decrease of the fluorescence count rate, owing to the time spent in this long lived state. Various techniques for laser-assisted deshelling have been proposed in the literature<sup>16</sup>. Such mechanisms should be eventually considered, in relation with a pulsed excitation scheme that will be required for true single-photon generation.

The experimental set-up is depicted in Fig. 1. It is based upon a home-made scanning confocal microscope. The green line ( $\lambda = 514$  nm) of a  $\text{Ar}^+$  cw laser is focussed on the sample by a high numerical aperture (1.3) immersion objective (Nachet 004279). A PZT-mounted mirror located just before the objective allows for a x-y scan of the sample, and fine z-scan is obtained using another PZT. The fluorescence (wavelength between 637 and 800 nm) is collected by the same objective and separated from the excitation laser by a dichroic mirror. High rejection rate ( $10^{15}$ ) long pass filters are used to removed any green or blue light. The confocal operation is achieved by imaging the sample onto a  $50\mu\text{m}$  pinhole with a magnification of 100. In order to investigate the fluorescence intensity correlation we use an Hanbury-Brown and Twiss setup with two avalanche photodiodes (EG&G, model SPCM-AQR 13) and a 50/50 beamsplitter. IR filters and pinholes are set in front of each detector to prevent optical cross-talk between them<sup>17</sup>. The time delay between pulses from the two photodiodes is converted by a time to amplitude converter (TAC) into a voltage amplitude, that is digitalized by the data acquisition card of a computer. A delay of 40 ns in one TAC input allows the measurement of the intensity correlation for negative time.

Fig. 2 shows a scan of the sample. Individual bright spots corresponding to N-V centers appear clearly on the scan, with an apparent lateral resolution of 500 nm. However, we note that the scan has been obtained at a depth of 10  $\mu\text{m}$  below the diamond surface, where the spherical aberration caused by the diamond-oil interface is not negligible (the refractive index of diamond is  $n = 2.4$ ). An evaluation done using a lens-design software indicates that only 20% of the emitted light is actually in the main peak, while about 80% is spread out in a halo which is about 3  $\mu\text{m}$  in diameter. Similar aberrations appear on the focused laser beam. For an input power of 15 mW, the typical signal from an individual center is  $S_{det} = 2000 \text{ s}^{-1}$  on each photodiode. The evaluated overall detection efficiency is  $\eta_{tot} = 0.0014$ . It can be split in  $\eta_{geom} = 0.08$  (geometrical collection angle),  $\eta_{ab} = 0.2$  (aberrations due to the diamond-oil interface),  $\eta_{opt} = 0.25$  (optical transmission),  $\eta_{BS} = 0.5$  (beamplitter), and  $\eta_{det} = 0.7$  (detector quantum efficiency). The inferred emission rate from the center is thus of  $S_{det}/\eta_{tot} = S_{em} = 1.4 \times 10^6 \text{ s}^{-1}$ . The difference between  $S_{em}$  and the fully saturated value  $S_{rad} = 9 \times 10^7 \text{ s}^{-1}$  is attributed to the joint effects of shelving, and of a non-saturated excitation. It can also be seen from Fig. 2 that the peaks appear above a background level with a typical value  $B = 4000 \text{ s}^{-1}$ . When increasing the incident laser power (in the range 2-20 mW), a saturation behaviour appears on the signal fluorescence, while the background level increases linearly. The intensity correlation from the signal and from the background also behave differently, as it will be discussed in more detail below.

The raw coincidences  $c(t)$  (right axis) and the correlation function  $g^{(2)}(t)$  (left axis) are represented in Fig. 3. There  $t$  is the delay between two joint photodetection events. In order to have a good statistics on the correlations, the typical data acquisition time  $T$  is a few hours. A slow (8 s response time) x-y-z computerized servo-lock was used to maintain the fluorescence on the maximum of the N-V center under study. Short-term and long-term drifts of the laser intensity are less than 10%.

For a Poissonian light source the coincidence rate (in  $\text{s}^{-1}$ ) in a time bin of width  $w$  is  $N_1 N_2 w$ , where  $N_{1,2}$  are the count rates on each detector. The raw coincidence number  $c(t)$  is normalized to the one of a Poissonian source according to

$$C_N(t) = c(t)/(N_1 N_2 w T) . \quad (1)$$

By stopping the scan between several centers (typically 1  $\mu\text{m}$  away from a center of interest), we have checked experimentally that the normalized coincidence rate from the background light is flat and equal to unity. We thus make the reasonable assumption that the photons emitted from the background are uncorrelated with the photons emitted from the center at the same location. This allows us to correct for the random coincidences caused by the background light and obtain the  $g^{(2)}(t)$  correlation function of the N-V center :

$$g^{(2)}(t) = (C_N(t) - (1 - \rho^2))/\rho^2 \quad (2)$$

where  $\rho = S/(S + B)$  is related to the signal to background ratio, which is measured independantly in each experimental run. After this subtraction procedure, the  $g^{(2)}(t)$  correlation function shown in Fig. 3(b) goes to zero for  $t = 0$ . This is consistent with our claiming that there is only a single N-V center in the fluorescence peak selected in Fig. 2. In the case of the presence of  $n$  spatially unresolved centers, the value of the zero-time antibunching is  $1 - 1/n$ . Experimental runs with  $g^{(2)}(0) > 0$  were also commonly observed, and are typical of the brightest spots seen

on Fig. 2. In Fig. 3(b), the non-unity value of  $g^{(2)}(t)$  for  $t > \tau$  is the bunching effect<sup>8,13,15</sup> owing to the presence of a metastable singlet state<sup>16</sup>. We have observed experimentally that this bunching effect depends on the laser power, indicating that the 514 nm excitation light may also contribute to deshelling the metastable state. A quantitative study of these effects is under way.

In the present experiment the background level ( $\rho = 0.34$ ) is too high for an efficient use in quantum cryptographic systems. Spectrographic analysis of the fluorescence light reveals that significant improvement should be obtained by optimizing the spectral filters and dichroic mirrors, in order to eliminate stray fluorescence around 600 nm which does not arise from the NV center. Preliminary measurements show that the fraction of useful signal  $\rho$  can then reach values above 0.8. Other improvements can also be expected from a non-aberrating collection optics, from truly fluorescence-free immersion oil, and from an optimized choice of the diamond sample.

As a conclusion, we have observed photon antibunching from a very simple set-up involving a diamond crystal at room temperature. The total absence of photobleaching allowed us to lock the laser beam on a single center during several hours. Colored centers are included in a solid matrix, easy to handle, and they appear as good candidates for realizing single photon sources for quantum cryptography. Ultimate efficiency for such a source should be obtained by coupling the emitting dipole to a microcavity, in order to emit the light in a single mode<sup>9</sup>. We point out however that a possible alternative for achieving overall efficiencies in the range 10 – 20% may be the use of optimized wide-aperture collection optics.

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Fig. 1. Experimental set-up. The sample fluorescence is excited and collected using a confocal microscope set-up. The intensity correlations are measured using two avalanche photodiodes, a time to amplitude converter and a multichannel analyzer.

Fig. 2. (a) Confocal microscopy raster scan ( $5 \times 5 \mu\text{m}^2$ ) of the sample performed about  $10 \mu\text{m}$  below the diamond surface. The size of a pixel is  $60 \text{ nm}$ . The integration time per pixel is  $32 \text{ ms}$ . The laser intensity impinging on the sample is  $15 \text{ mW}$ . (b) Line scan along the dotted line in (a). The data is shown together with a gaussian fit, which is used to evaluate the signal and background levels. Here we obtain  $\rho = S/(S + B) = 0.34$

Fig. 3. Normalized correlation function  $g^{(2)}(t)$  corrected for the random coincidences from the background ( $\rho = 0.34$ , see text). The data corresponds to the center circled in Fig. 2(a). The actual number of coincidences is indicated on the right. The time bin is  $w = 1 \text{ ns}$ , the integration time is  $T = 11450 \text{ s}$ , and the single count rates are  $N_1 = 5780 \text{ s}^{-1}$ ,  $N_2 = 5990 \text{ s}^{-1}$ . The full line is an exponential fit to the data, using the model described in ref.<sup>8</sup>.

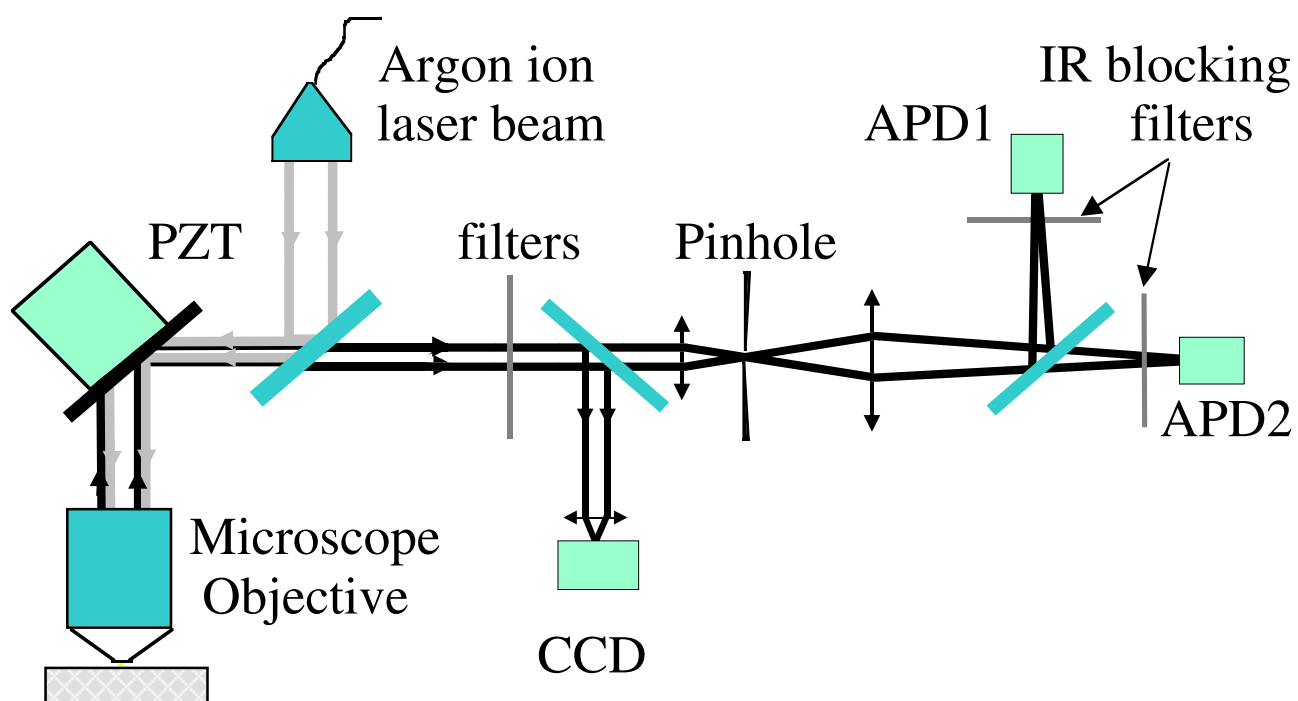


Fig.1 Brouri et al (Opt. Lett.)

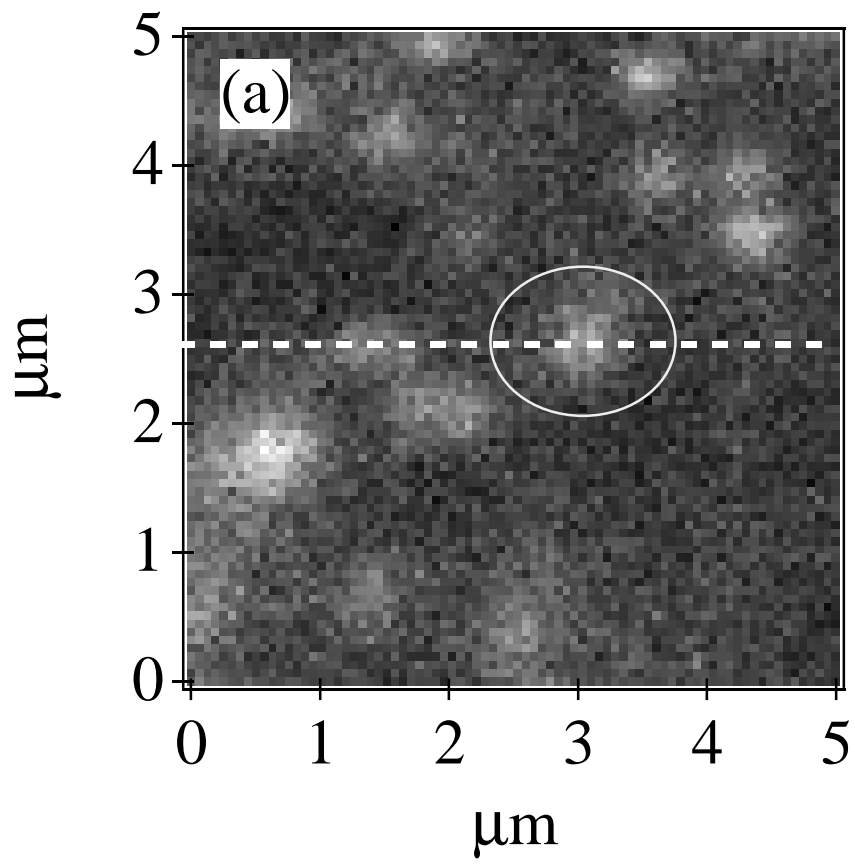


Fig.2 (a) Brouri et al (Opt. Lett.)

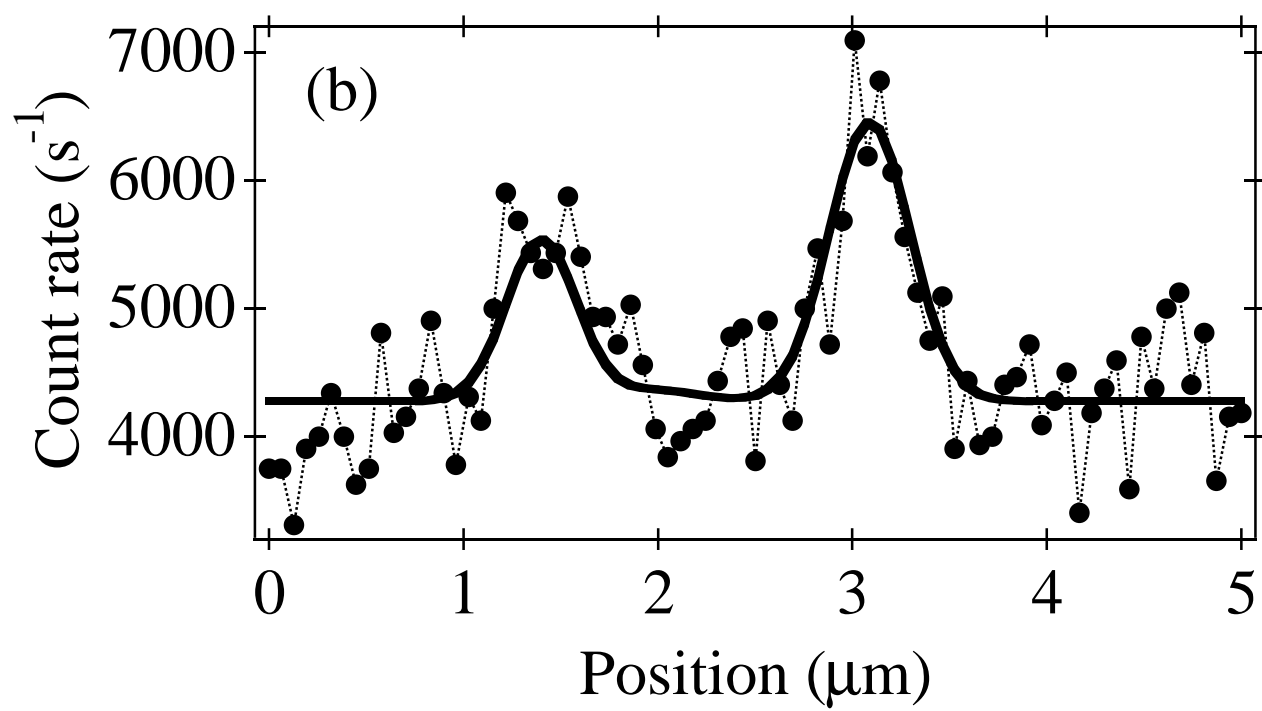


Fig.2 (b) Brouri et al (Opt. Lett.)

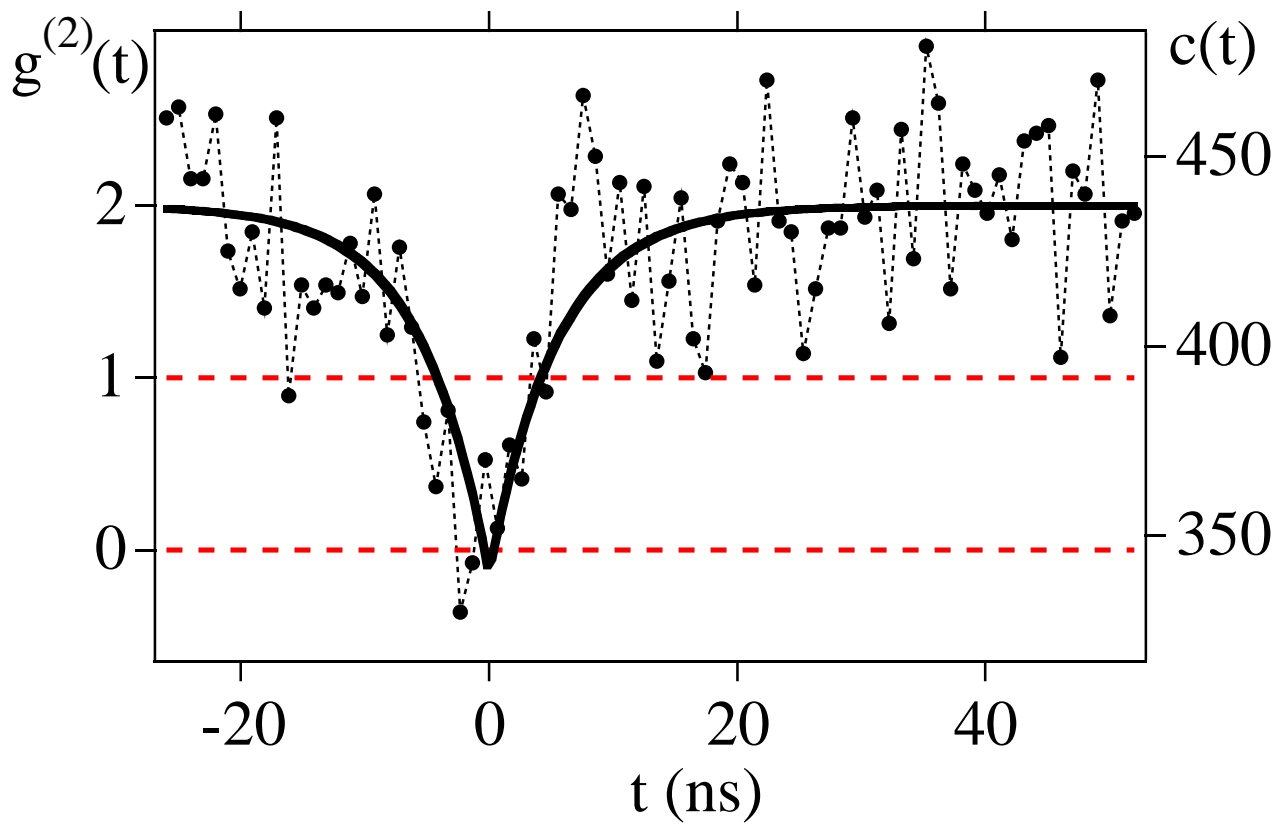


Fig.3 Brouri et al (Opt. Lett.)